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An Eclectic Journey Through Experimental Nuclear Physics, or How I Learned to Stop Worrying and Love Nuclear Data

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An Eclectic Journey Through Experimental Nuclear Physics, or How I Learned to Stop Worrying and Love Nuclear Data

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Abstract. In this paper, I illustrate how the ability to rapidly access the broad range of nuclear data has facilitated my research in fields from searches for double beta decay, to measurements of astrophysical reaction rates, to issues in homeland security. In doing this, I hope to persuade even the skeptics that for the benefit of the broad scientific community, it is imperative that the outstanding work of the nuclear data community continue.

INTRODUCTION

The databases that are compiled, evaluated, and disseminated by the national and international nuclear data programs are invaluable resources for basic physics research, engineering, medicine, and a host of applied activities. Sometimes, this fact is not fully appreciated by either our colleagues or by the funding agencies that pay for the work that is required to produce and maintain this information.

DOUBLE BETA DECAY

An important background source in double beta-decay experiments is due to the presence of long-lived cosmogenic nuclides in the source materials. Cosmic-ray induced spallation reactions on medium to large mass nuclei can produce a wide range of radioisotopes. ^{60}Co is a particularly challenging case because of its large total decay energy (2.824 MeV), its β-decay endpoint energy (0.318 MeV), and its coincident γ rays at 1.173 and 1.332 MeV [1]. If ^{60}Co is present in an active detector, then its decays can produce events from several different combinations of the β and γ rays. We measured the cross sections for the production of ^{60}Co in ^{nat}Mo and ^{nat}Te at the proton energy of 1.85 GeV. The irradiation was performed in 1992 at LBNL's Bevalac facility. The targets were γ-ray counted immediately after the irradiation and the production cross sections for many isotopes (although not for ^{60}Co) were determined [2]. Eleven years later, these targets were gamma-ray counted at LBNL's Oroville Dam Low Background Facility. The spectrum obtained in 2003 from the Te target is shown in Fig. 1.

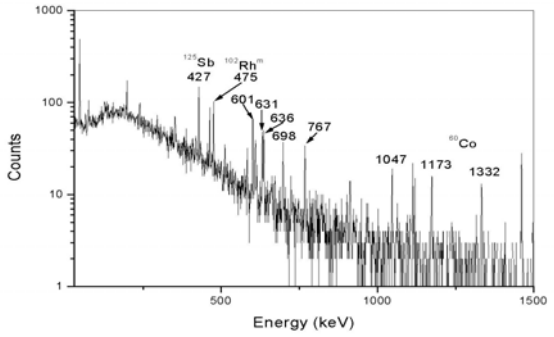

FIGURE 1. γ-ray spectrum observed in 2003 from the ^{nat}Te target bombarded in 1992 with 1.85-GeV protons.

The characteristic 1173- and 1332-keV lines from ^{60}Co are clearly observed, along with lines from two other long-lived products, ^{102}Rh^{m} and ^{125}Sb. The production cross sections for these latter two nuclides were determined in our original study of these activated targets. Based on our earlier results [2] and the half-lives listed in ENSDF for these two isotopes, I expected that the ratio of activities of these two isotopes should now be 2.31. However, what I observed was a ratio of 1.42 ± 0.13. In looking at the

ENSDF file for A=102, I noted that the half-life value listed for $^{102}Rh^{m}$ was shown as ~2.9 years. I wondered if in the time since the literature cutoff for this evaluation, any new measurements of this half life had been made. I went online to check NSR and soon found a reference to a publication by Shibata *et al.* [3]. With another few mouse clicks, I retrieved this paper. I soon found that Shibata *et al.* found a value of 3.742 $\pm$ 0.010 years for the half-life of $^{102}Rh^{m}$. While this may not seem like a significant difference from 2.9 years, after 11 years, it has a considerable effect. Using the half-life value of Shibtata *et al*., I found that the expected activity ratio should now be 1.32, in good agreement with my measured value. This story illustrates the importance of half-life measurements as well as the usefulness of having easy and quick access to ENSDF, bibliographic databases such as NSR and the ability to directly retrieve published articles.

DETECTING FISSILE MATERIAL IN SEAGOING CARGO CONTAINERS

Approximately 6 million seagoing cargo containers come into U.S. ports each year. A major security issue is to develop a means to scan such containers for the presence of fissile material. About two years ago, Prof. Stan Prussin and I were both spending sabbatical visits at Lawrence Livermore National Laboratory (LLNL). The group we were in was investigating this problem and had suggested looking for beta-delayed neutrons emitted from fission fragments following active interrogation. We thought about this idea for a while, but soon became convinced that because of the small emission probability for beta-delayed neutrons, their low energies, and the presence of hydrogen in many cargo scenarios, the likelihood of detecting fissionable material using this technique is low.

We then spent a couple of weeks studying various nuclear databases to look for a more robust signature. Using the tabulated yields of fragments produced by the thermal neutron induced fission of both ^{235}U and ^{239}Pu [4], and the online version of the *Table of Radioactive Isotopes* [5], we found that beta-delayed gamma rays with energies above 3 MeV are emitted by fission fragments with approximately 10 times higher intensity than beta-delayed neutrons. This higher intensity combined with their large penetrating power suggested to us that these high-energy gammas might be an easier signal to detect. These gamma rays are emitted by about 20 different fission products whose half-lives vary from about 0.5 to 250 seconds. Furthermore, 3 MeV is above the energy of essentially all natural background radiation. We also found that there appeared to be few, if any, other radioisotopes that would be produced by neutron activation that would emit gamma rays above 3 MeV.

Our hosts at LLNL were a bit skeptical about our ideas, so we decided to perform some proof-of-principle experiments at Lawrence Berkeley National Laboratory's 88-Inch Cyclotron. There we produced neutrons by bombarding a thick Be target with 16-Mev deuterons. The neutrons were thermalized in a moderator constructed of iron and polyethylene. Targets were shuttled from the irradiation point inside the moderator to a remote shielded counting station where we had both a high-resolution germanium detector and a 30x30x5-cm plastic scintillator. We irradiated each target for 30 seconds and then counted for 30 seconds in ten sequential 3-second time bins. Figure 2 illustrates the spectrum observed in the germanium detector from the bombardment of 0.568 grams of ^{239}Pu and 115 grams of steel. The time dependence of the high energy gamma rays is shown in the inset. The energy and temporal distribution of the high-energy gamma rays from the fission of ^{235}U are very similar to those of ^{239}Pu, but their intensity per fission is approximately 3 times larger.

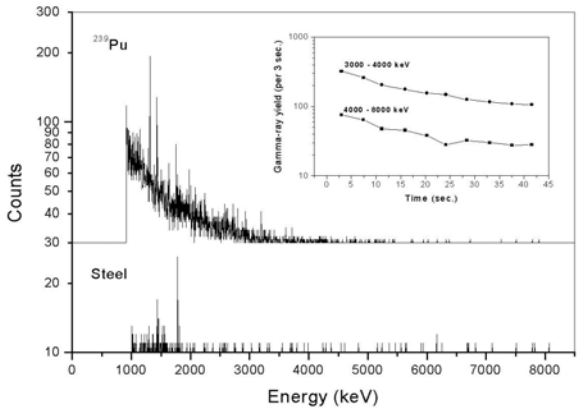

FIGURE 2. γ-ray spectrum and temporal variations observed following the neutron irradiation of 0.568 grams of ^{239}Pu and 115 grams of steel [6].

Results similar to those from steel were also found from irradiations we performed on aluminum, wood, polyethylene, and sandstone. In particular, from all of the non-fissile materials we observed a small number of low-energy gamma rays produced by the decays of reasonably long-lived nuclides. On the other hand, the spectra from ^{235}U and ^{239}Pu exhibit fairly strong emission of gammas that extend up to approximately 6 MeV that decay with an effective half-life of

approximately 25 seconds. These two features, large numbers of gamma rays above 3.0 MeV and a short effective half life, are unique signatures of fissile material [6].

Despite its relatively poor energy resolution, we were able to obtain similar identification of fissile material using the data from the plastic scintillator. In order to scan full size cargo containers in a reasonable time, high efficiency detectors will be required. Thus, we are now investigating the use of large plastic or liquid scintillators as a means to detect the high-energy gamma rays following active interrogation. For more details on this subject, see the contribution by S. Prussin in these proceedings.

HALF LIFE OF $^{108}Ag^{m}$

Sometimes, in the course of doing their work, nuclear data evaluators encounter situations where results of different measurements give conflicting results. The half-life of the long-lived isomer $^{108}Ag^{m}$ is an example of this type of situation. As can be seen in Table 1, values ranging from 127 to 438 years have been reported. Because of these very discrepant results, it is not appropriate to simply quote an average as the "best value" for this half life. This situation prompted Edgardo Browne and I to undertake a remeasurement of the half life of $^{108}Ag^{m}$.

We obtained a source of $^{108}Ag^{m}$ from Richard Helmer and mounted it together with sources of ^{44}Ti and ^{133}Ba near a 3.6-cm diameter by 1.3-cm thick planar germanium detector. Data is collected automatically using an ORTEC PC-based data acquisition system. Spectra are accumulated in one-week intervals, written to disk, cleared, and then started again. We have collected data for more than one year. One of the spectra collected during one week of counting is shown in Figure 3.

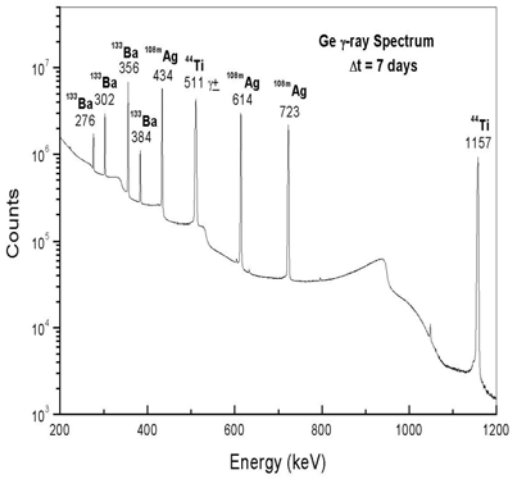

FIGURE 3. γ-ray spectrum obtained in 7 days of counting the mixed source of ^{44}Ti, $^{108}Ag^{m}$, and ^{133}Ba.

The half life of ^{133}Ba is well known and serves as a reference in our measurement. In order to establish that our experiment is working properly, we extract the half-life of ^{44}Ti by analyzing the ratio of the number of counts in the 356-keV ^{133}Ba peak to the number in the 1157-keV ^{44}Ti peak. The results of this analysis are shown in Figure 4. The value we obtain for the half life of ^{44}Ti of 57.6 $\pm$ 1.0 years is in reasonably good agreement with the recently evaluated result of 59.6 years [11].

We then analyze the ratio of the 356-keV line from ^{133}Ba to that of the sum of the areas of the 434-, 614-, and 723-keV lines from $^{108}Ag^{m}$ to obtain the half life of $^{108}Ag^{m}$. The results of this analysis are shown in Figure 5. Our preliminary result for this half life is 440 $\pm$ 84 years. This result is in very good agreement with the most recently reported result of Schrader [10]. We are continuing to collect more data in order to reduce the statistical uncertainty in our result.

TABLE 1. Reported Values of $^{108}Ag^{m}$ Half-life

Half-Life (y)	Method	Duration	Reference
127 (21)	Specific Activity		Harbottle [Ref. 7]
310 (132)	Specific Activity		Vonach *et al.* [Ref. 8]
418 (15)	Decay	10 y	Schotzig *et al.* [Ref. 9]
437.7(88)	Decay	22 y	Schrader [Ref. 10]

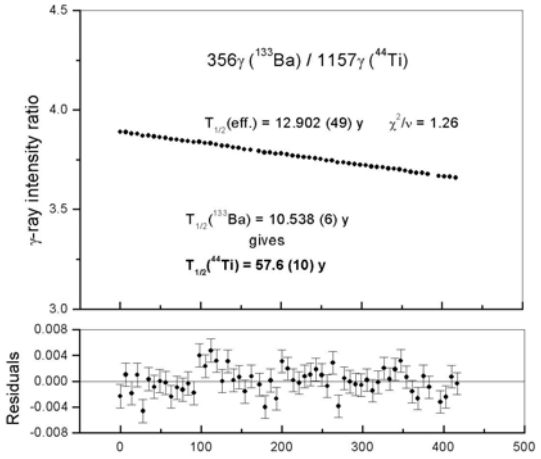

Figure 4. Analysis of the ratio of the area of the 356-keV ^{133}Ba line to that of the 1157-keV line from ^{44}Ti.

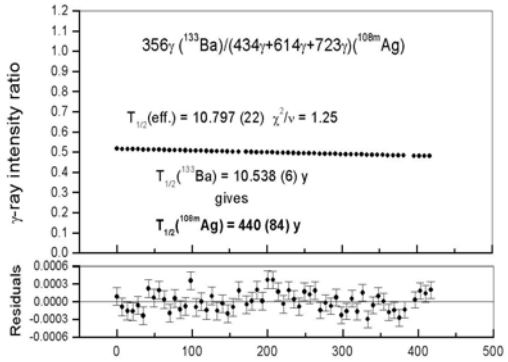

Figure 5. Analysis of the ratio of the area of the 356-keV ^{133}Ba line to the sum of the areas of the 434-, 614-, and 723-keV lines from ^{108}Ag^{m}.

(α,γ) CROSS SECTIONS

The nucleosynthesis of many nuclides is the result of charged-particle induced reactions that take place in stars. While there is an enormous literature on proton-induced reactions, there is surprising little published data for alpha-induced reactions. In fact, by looking at the EXFOR database [12], one finds that (α,γ) cross section data have been reported for only 12 target nuclei with A > 60. Thus, a group at LBNL decided to perform several new measurements using the stacked foil activation technique. As reported at this meeting, Basunia *et al.* [13] measured cross sections for the ^{63}Cu(α,γ)^{67}Ga reaction. A comparison of these measurements with the results of a Hauser Feshbach calculation is shown in Figure 6. Baglin *et al.* [14] studied the ^{107}Ag(α,γ)^{111}In reaction. Results from this study are shown in Figure 7. As had been seen in the limited number of previous such investigations, the Hauser Feshbach calculations do a reasonably good job matching the experimental data for the lighter mass target. However, there is a much larger discrepancy for the heavier target. Additional experiments are planned to further investigate this issue.

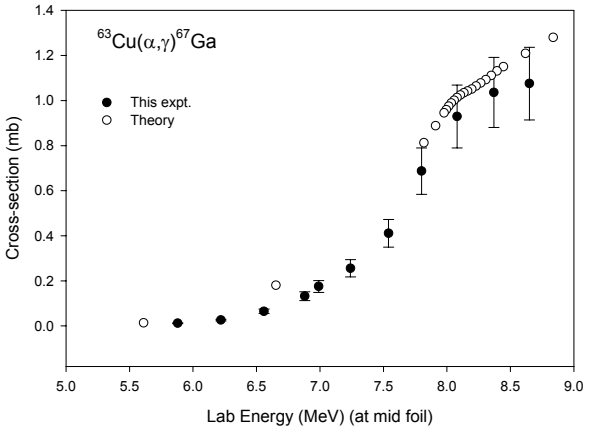

Figure 6. Experimental and theoretical cross sections for the ^{63}Cu(α,γ)^{67}Ga reaction from Basunia *et al.* [13].

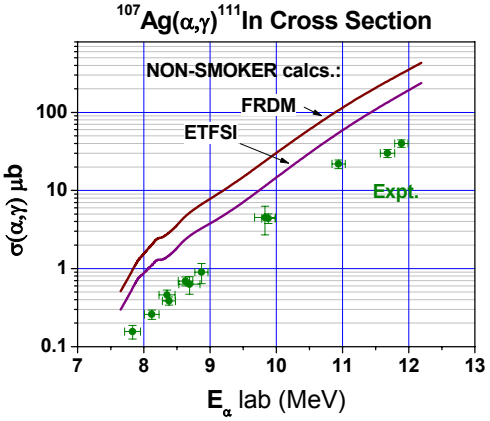

Figure 7. Preliminary cross section data for the ^{107}Ag(α,γ) reaction compared with NON-SMOKER calculations from Baglin *et al.* [14].

CONCLUSIONS

I hope that in this paper, I have illustrated how having quick and easy access to evaluated nuclear data is a major asset to research in basic and applied science. Evaluation also points out needs for future experimental activities. Sometimes the evaluators themselves become experimenters in order to address these needs. Thus, I know that I am not alone in believing that the work of the national and international programs in nuclear data compilation, evaluation, and dissemination need to be continued and supported.

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REFERENCES

1. E. Browne, R. B. Firestone, V. S. Shirley, *Table of Radioactive Isotopes*, John Wiley, New York 1986.
2. D. W. Bardayan *et al.*, Phys. Rev. C **55**, 820 (1997).
3. M. Shibata *et al.,* Appl. Rad. & Isot. **49**, 1481 (1998).
4. T. R. England, B. F. Rider, LA-UR-94-3106; ENDF-349.
5. http://ie.lbl.gov/toi.
6. E. B. Norman *et al.,* Nucl. Instr. & Meth. A **521**, 608 (2004); Nucl. Instr. & Meth. A **534**, 577 (2004).
7. G. Harbottle. Radiochim. Acta **13**, 132 (1970).
8. H. Vonach, M. Hille, P. Hille. Z. Physik **227**, 381 (1969).
9. H. Schrader, Appl. Rad. and Isotopes **60**, 317 (2004).
10. U. Schotzig, H. Schrader, K. Debertin. Proc. Inter. Conf. Nuclear Data for Science and Technology, Julich, Germany (1992) p. 562.
11. J. A. Cameron, B. Singh, Nucl. Data Sheets **88**, 299 (1999).
12. www.nndc.bnl.gov/exfor.
13. S. Basunia *et al.*, these proceedings.
14. C. M. Baglin *et al.*, these proceedings.